

# SUBSTANTIAL NUCLEAR ORDERING IN SOLID $\text{He}^3$ <sup>†</sup>

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Evidence for a substantial nuclear ordering in solid bcc  $\text{He}^3$  at melting pressure has been inferred from solid entropies  $s_S$  deduced from measurements of the slope of the melting curve for a liquid-solid mixture cooled by adiabatic compression. At 3.4 mK we find experimentally that  $s_S/R = 0.83 \ln 2$ . It is likely that much lower temperatures and solid entropies are obtained during parts of the experiments. The possibility for bulk nuclear polarization is also discussed.

It is possible to convert an ordered Fermi liquid to a solid with ordered nuclear spins.<sup>1</sup> More specifically, highly ordered liquid  $\text{He}^3$  may be produced readily at temperatures of 10 to 20 mK, a temperature region easily attained by dilution refrigeration, since liquid  $\text{He}^3$  is a Fermi liquid with an effective Fermi temperature much greater than 20 mK. Isentropic conversion of liquid to solid should then produce a highly nuclear-spin-ordered solid. Quantitatively, the ordering of the solid may be measured by its entropy. In this work we determined the solid entropy by measuring the slope  $dP/dT$  of the melting curve and using the Clausius-Clapeyron equation

$$\frac{dP}{dT} = -\left(\frac{s_S - s_L}{v_L - v_S}\right), \quad (1)$$

where  $s_S$ ,  $v_S$  and  $s_L$ ,  $v_L$  are the molar entropy and volume on the melting curve for the solid and liquid phases, respectively. Alternatively, nuclear ordering could be observed by forming the solid in a strong magnetic field and measuring the bulk nuclear magnetic moment. This has not been done in the present experiments, but work along these lines is in progress.

At temperatures below a few tenths of a kelvin the entropy of solid  $\text{He}^3$  results from disorder in the nuclear-spin system. In zero external field, the spins may be described by an effective spin

Hamiltonian<sup>2</sup>

$$\mathcal{H} = -J \sum_{ij} \vec{I}_i \cdot \vec{I}_j, \quad (2)$$

where  $J$  is a parameter, and the sum is usually assumed to be over nearest neighbors. Using data of Anderson, Reese, and Wheatley<sup>3</sup> we showed<sup>4,5</sup> that  $J < 0$ , in agreement with recent measurements by Pipes and Fairbank<sup>6</sup>; Kirk, Osgood, and Garber<sup>7</sup>; and Sites, Osheroff, Richardson, and Lee.<sup>8</sup> Hence in zero external field, antiferromagnetic nuclear ordering is to be expected at low enough temperatures.

In recent work we demonstrated<sup>4</sup> that it is possible to cool a two-phase mixture of liquid and solid  $\text{He}^3$  mechanically to temperatures below 3 mK. However, we did not determine the resultant nuclear ordering. In some preliminary work we measured the nuclear-spin diffusion coefficient  $D$  in a solid formed by adiabatic compression. We found that, at the lowest temperatures,  $D$  had decreased to about  $\frac{2}{3}$  of its temperature-independent value at higher temperatures.<sup>9</sup> This result indicated that something interesting was happening in the nuclear-spin system. However, we interrupted the diffusion measurements in favor of the melting-curve measurements since these have the double advantage of yielding a direct measurement of solid entropy and of being much less sensitive to thermal disequilibrium in the solid nuclear-spin system.

From 0.3 K down to about 30 mK the solid molar entropy is essentially  $R \ln 2$  (corresponding to random orientations of the spin- $\frac{1}{2}$  nuclei) as deduced from the liquid entropy at the minimum of the melting curve<sup>10</sup> and from solid heat-capacity measurements<sup>11</sup> at a few hundredths of a kelvin. The liquid entropy at the melting curve at low temperatures may be calculated self-consistently using the liquid heat capacity at 27.0 atm given by Abel, Anderson, Black, and Wheatley<sup>12</sup> extended to the melting curve, using the expansion coefficient and compressibility given by Anderson, Reese, and Wheatley.<sup>13</sup> Since the difference  $v_L - v_S$  between the liquid and solid molar volumes is known at low temperatures from the work of Scribner, Panczyk, and Adams,<sup>14</sup> measurement of  $dP/dT$  along the melting curve allows the solid entropy to be determined from Eq. (1). Furthermore, the temperature calibration can be, and in this work is, determined by taking the solid molar entropy at higher temperatures to be  $R \ln 2$ .

The low-temperature portion of our apparatus is shown in Fig. 1. Details concerning its operation will be given in a paper currently in preparation. The  $\text{He}^3$  is compressed via the flexible-walled section by pressurizing liquid  $\text{He}^4$ .<sup>15</sup> The epoxy appendix to the flexible section contains the thermometer and strain gauge for the measurements to be presented here. The cell is pre-cooled by mounting it with the upper portion immersed in the mixing chamber of a continuously operating dilution refrigerator.<sup>16</sup> At low temperatures thermal isolation is provided by the thermal boundary resistance between the helium and the container walls.

The pressure of the  $\text{He}^3$  is measured with a beryllium-copper strain gauge<sup>14</sup> having a sensitivity of  $10^{-4}$  atm, which is calibrated at 1 K with only liquid in the cell. A magnetic temperature  $T^*$  is determined by measuring the 17-Hz susceptibility of powdered cerium magnesium nitrate (CMN) uniformly packed in a right-circular cylinder with diameter equal to height. For CMN we assume<sup>17, 18</sup>  $T = T^* + \Delta$  down to 3 mK with  $|\Delta|$  less than about 0.2 mK. We have taken  $\Delta = 0$  in all computations. The slope and intercept of the straight line relating mutual inductance and  $T^{*-1}$  were adjusted as indicated above using Eq. (1) in the range 20 to 38 mK to give a best fit of the data to the calculated curve of  $dP/dT$  vs  $T$ . The resultant melting curve is in general agreement with that of Ref. 14.

Averaged results obtained during the slow

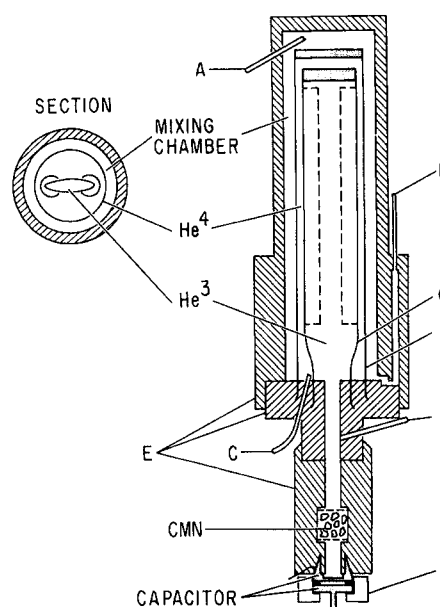


FIG. 1. Schematic of compression cell mounted in the mixing chamber of the continuous dilution refrigerator. A, inlet to mixing chamber. B, outlet of mixing chamber. C,  $\text{He}^4$  inlet. D,  $\text{He}^3$  inlet. E, heat-curing epoxy resin. F, nylon. G, flexible part of  $\text{He}^3$  cell consisting of a flattened 9.52-mm-o.d.  $\times$  0.41-mm-wall 70-30 cupronickel (CuNi) tube with slit 4.76-mm-o.d.  $\times$  0.25-mm-wall CuNi tubes for springs. H, outer wall of  $\text{He}^4$  space consisting of a 15.9-mm-o.d.  $\times$  0.41-mm-wall CuNi tube. CMN, powdered cerium magnesium nitrate. CAPACITOR, beryllium copper plates separated by 0.025-mm-thick Mylar ring.

warm-up following compression are shown for five runs in Fig. 2. Owing to uncertainties in the temperature scale and/or problems with thermal equilibrium, lower-temperature data were dis-

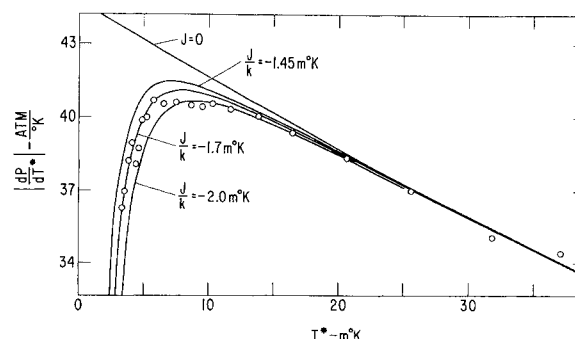


FIG. 2. The slope of the melting curve versus temperature as determined experimentally and as theoretically determined for several values of the parameter  $J$  in the Hamiltonian  $\mathcal{H} = -J \sum \vec{I}_i \cdot \vec{I}_j$ . The theoretical curves assume  $T = T^*$ . The experimental points are averages of five warming runs.

carded. Also shown on the figure are several curves calculated, for several values of the interaction parameter  $J$  in Eq. (2), by means of the ten-term high-temperature expansion for a bcc Heisenberg antiferromagnet and reported by Baker, Gilbert, Eve, and Rushbrooke.<sup>19</sup> According to the  $(\partial P/\partial T)_V$  measurements of Panczyk *et al.*,<sup>20</sup> a value of  $|J|/k = 1.44$  mK is to be expected for the solid at its melting pressure near  $T=0$ . We do not believe that our measurements are accurate enough to allow  $J$  to be determined well. However our results, favoring a somewhat higher value of  $|J|$  than that given by Panczyk *et al.*,<sup>20</sup> are sufficiently close to the theoretical curve, calculated using their parameter, to give us confidence in the general correctness of our data.

There is already substantial nuclear ordering at the lowest temperatures for which we have  $dP/dT$  measurements. At 3.4 mK we calculate from our  $dP/dT$  data and Eq. (1) that  $s_S = 0.58R = (0.83) \times R \ln 2$ . This is independent of any theoretical model. To give some perspective to this value for the entropy, consider a different physical system which consists of independent spin- $\frac{1}{2}$  particles in a magnetic field. A molar entropy of  $0.58R$  corresponds to a polarization of 46% in such a system.

Although our magnetic thermometry is no longer adequate below 3 mK, the melting pressure should be a meaningful thermometer. At 3 mK, the observed melting pressure is 33.91 atm while our highest observed pressure is 33.95 atm. Thompson and Meyer<sup>21</sup> have calculated the melting curve to  $T=0$  for  $J/k = -1.2$  mK and for  $J=0$ . We have performed a similar calculation of the melting curve for several values of  $J$  using our calculation for  $s_L$ ,  $v_L - v_S$  from Ref. 14,  $s_S$  calculated using the expansion in Ref. 19 above  $T_N (= 1.374|J|/k)$ , and the spin-wave  $T^3$  expression in Ref. 14 at low temperatures with a smooth graphical connection through the intermediate region. Starting from 5.0 mK and 33.831 atm, a point on our observed melting curve, we calculate at  $T=0$  a melting pressure of 33.967 atm for  $J/k = -1.2$  mK, 33.952 atm for  $-1.44$  mK, and 33.935 atm for  $-1.7$  mK. For  $J/k = -1.44$  mK, as determined in Ref. 20, we calculate a melting pressure of 33.910 atm at 3 mK in agreement with observation. Our maximum pressure corresponds, for this  $J$ , to about 1.5 mK and to  $s_S = (0.1)R \ln 2$ . These figures refer to the liquid-solid interface.

When the solid  $\text{He}^3$  is formed in a magnetic field,<sup>22</sup> the effective nuclear-orienting field  $H_{\text{eff}}$

is less than the applied field  $H_{\text{appl}}$  as a result of the effective antiferromagnetic spin interaction. Using  $J/k = -1.5$  mK we estimate

$$H_{\text{eff}} \approx H_{\text{appl}} - 4 \times 10^{-4} P_n \text{ G}, \quad (3)$$

where  $P_n$  is the nuclear polarization, defined to be positive. If liquid  $\text{He}^3$  is converted isentropically to solid in an applied field of 70 000 G, a 76% nuclear polarization (corresponding to  $|g_N \beta_N| H_{\text{eff}} / 2kT = 1$ ) will be attained at a temperature of 3 mK. Starting with liquid at 20 mK, only 22% of the  $\text{He}^3$  needs to be converted to solid to achieve this result. Furthermore, by suitable heating<sup>8</sup> the solid can be formed where required. Whether or not such a high polarization can be attained in practice depends at least on mechanical irreversibilities in the device. In parts of our present work to be presented elsewhere, we find such irreversibilities small. A more serious problem appears to be the attainment of thermal equilibrium within the solid nuclear-spin system itself.

A more detailed account of our work on the melting curve is in preparation. We wish to acknowledge here the important contributions made to measurements of the melting curve, preliminary to those presented here, by Dr. Ralph Rosenbaum and Professor Olli Lounasmaa.

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